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# Beam-Foil Spectroscopy - Quo Vadis?

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**Abstract.** Beam-foil spectroscopy after 45 years: what has been realized of the promises, what is the state of the art, what is the status of the field, what present atomic physics problems should the technique be applied to, where can it be done? Will it be done?

Electronic excitation and ionization of atoms (including beam-foil excitation and ionization) - Atomic lifetimes - Oscillator strengths - Visible and ultraviolet spectra

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## 1. Introduction

Beam-foil spectroscopy (BFS) and its derivatives (for example, beam-gas spectroscopy, recoil-ion spectroscopy, and some experiments at heavy-ion storage rings) are atomic physics techniques that exploit fast ion beams as are provided by ion accelerators which usually have been set up for nuclear physics experiments in the first place. This starting condition holds many clues to the stage setting on which BFS has evolved, grown, bloomed, and thereafter largely faltered. For the purposes of nuclear science, energetic ion beams have been produced by ever more powerful ion accelerators so that the ions could be targeted at specific atomic species (usually in the form of a thin foil or deposited on a thin foil backing). If the energy was high enough, the nuclei of projectile ions and target atoms would interact and nuclear reactions might take place. The thin-foil target design enables the reaction products to suffer minimal energy loss on their way to a particle or  $\gamma$ -ray detector. Apparently, in several accelerator laboratories people noticed

‡ Dedicated to the memory of Prof. S Bashkin (20 Jun 1923 - 1 May 2007)  
and in honour of Prof. L J Curtis (\* 4 Nov 1938)

that with strong ion beams there was also observable optical radiation. However, in only two nuclear physics laboratories (Manchester, England and in Iowa, USA) were there nuclear physicists (L Kay [1] and S Bashkin [2, 3, 4], respectively) who ventured to have a closer look at this phenomenon outside of nuclear physics. Both scientists drew ridicule and severe criticism from their faculty colleagues for their deviant pursuits. Kay felt forced to give up this new line of research [5], while Bashkin persevered after moving to the University of Arizona (Tucson) [6] and turned his new laboratory into a center for the development of the new technique [7]. Notably the visitors of the early generation (H J Andr , H G Berry, B Curnutte, J D sesquelles, I Martinson, R Schectman (who inspired L J Curtis), E Veje) carried accelerator-based atomic physics to their home institutions where they set up or expanded groups of their own. Bashkin died in 2007; all five of that early group of disciples have reached or passed retirement age by now, and it may not be surprising to see a similar life cycle coming to a conclusion of the field itself.

Bashkin avidly propagandized the promises of the new technique [4]. Although BFS was taken up in many accelerator laboratories, it was regularly done so as a side effort in a laboratory usually dedicated to nuclear physics, on accelerators that offered free capacity after their nuclear physics masters looked for higher energy machines in order to dig deeper into the nucleus. Atomic physics using accelerated ion beams was very suitable for university teaching and research, and a number of such groups were established.

While such second-hand usage of the accelerator facilities was a low-investment way for small atomic physics groups to obtain accelerator beam time, it left them out of the power to influence the machine development or have a major say on resource assignment. But when in the long run many of the post-world war II accelerator facilities were shut down, as nuclear physics moved on to higher ion beam energies, BFS lost the convenient access to local facilities and the field consequently shrank. This picture of US and European facilities was followed in Japan, India, and China. An exception are the heavy ion storage rings that have largely been built with nuclear physicists' accelerator expertise, but have a large fraction of their time dedicated to atomic and, increasingly, molecular physics. Some of this work can be seen as a straightforward extension of BFS.

Several Comments have reported on BFS work [8, 9, 10, 11], but the latest wide-ranging Comment is of 1989 and thus has appeared in a period in which internationally the level of BFS activities peaked and in which reaching the few-electron ions of uranium [23] indicated the end of the era of competition for this goal. Since then, more BFS work has been published than can be reviewed here (the four authors of those three previous Comments alone have authored or coauthored about 150 publications which pertain to BFS or beam-gas spectroscopy); some of the work has been summarized elsewhere [12, 13]. BFS work at small accelerators in this period has, for example, been pursued at the University of Toledo (OH, USA) and at the Tata Institute in Mumbai (India); medium-sized accelerators served BFS at (among other places) Argonne National Laboratory (U.S.A.), Lund University (Sweden), Universit  Laval

(Québec, Canada), Aarhus University (Denmark), University of Bochum (Germany), China Institute of Atomic Energy (Beijing, P. R. China), Tata Institute of Fundamental Research (Mumbai, India) and Nuclear Science Centre (New Delhi, India), while among larger accelerators almost the only machine occasionally still used for BFS was the Unilac at GSI Darmstadt (Germany) (earlier such large machines had included the Berkeley SuperHILAC and Bevalac as well as GANIL Caen (France)). Specialized reviews of lifetime measurements on Li- and Be-like ions [14], and the BFS offspring of lifetime measurements that are employing heavy-ion storage rings have been reviewed elsewhere [15, 16]. In the following I shall discuss selected BFS development highlights and achievements, and then expand on a few examples of persisting atomic physics problems for which BFS would be a most suitable tool.

## **2. Achievements**

Many of the technical principles and methods of accelerator-based atomic physics have been reviewed elsewhere (for example, in [17]). Here I try to highlight achievements of experiments that I consider interesting and serve as a basis to discuss the present possibilities and prospects of the field.

### *2.1. Excitation*

The ion-foil interaction takes place inside a solid, mostly with the electrons of the foil target (there are many more target electrons than target nuclei). Hence the excitation occurs in a high-density environment, denser than most other light sources and rivaled only recently by extreme power-density laser-produced plasmas. Meanwhile, by beam-foil interaction ions of all charge states have been produced, from negative ions [18, 19, 20, 21, 22] to bare uranium [23], a feat not matched (by far) by laser-produced plasmas. The fast-beam ions, after their high collision frequency encounters inside the high particle density target, travel on, and upon leaving the exciter foil they reach a low-density environment. BFS has studied the role of wake and convoy electrons and the influence of surface fields on the details of the electron cloud that accompanies the ions. These influences are of interest, but the overriding parameter is again the target density: it is so low (high vacuum) that the ions can be observed essentially without perturbations by further collisions. A somewhat lower-density target can be realized by replacing the foil by a differentially pumped gas cell or an open gas jet; at lower target densities, the average charge state of the emerging beam and the width of the charge state distribution is generally lower, similarly to the difference between foil and gas stripping in the high voltage terminal of a tandem accelerator. A gas target is less well localized than a foil target, which necessitates using the latter for experiments that achieve high time resolution by high spatial resolution along the ion beam. But there also are experiments in which the actual target density or the density profile is not of importance.

What does matter is the fact that high-density excitation leads to the population of high-lying levels, of multiply excited levels (several electrons not in the ground configuration), and of levels with high total angular momentum  $J$ . Some of the electron capture at the rear surface of the foil target also favours high- $n$  (principal quantum number  $n$ ), high- $\ell$  (angular momentum quantum number  $\ell$ ) levels. Such levels decay towards the yrast line of levels with a maximum value of  $\ell$  for a given value of  $n$ . On that line, radiative decays proceed by many stages of  $\Delta n=1$  transitions. High-lying levels on the yrast line can be quite long-lived, even in highly charged ions. In time-resolved measurements of decays of low-lying levels, cascade repopulation matters, and the superposition of components of the yrast decay chain both boosts the apparent level population (by the fast cascades from rather low-lying levels) and provides a long drawn cascade tail. This cascade tail has too many contributions to be treated by any multi-exponential analysis, but it can be approximated by a power law for practical purposes. A frequent finding is a time dependence of the yrast cascade proportional to  $t^{-1.5}$  (see refs. in [24]), and models of the level population mechanism have been based on such analyses. However, just as with attempts of finding general laws of the level population of (much more ‘individual’ in energy and angular momentum) lower lying levels, no simple population law has been found that would fit to many cases. Models and approximations abound. Extensive transport calculations for specific systems have also shown their value [25], but are not generally available. There may be resonance effects of electron binding energies such as with the work functions of the foil material (see references in [26]). However, it would seem futile to continue searching for a detailed description and parametrization of what is basically a statistical process which has eluded quantification by so many valiant attempts. In practice, if the cascades matter, they have to be measured or, at least, they have to be modeled with educated guesswork on the basis of a given atomic system with calculated level lifetimes and branch fractions.

The decay curve of a given atomic level reflects the lifetime of the level as well as its repopulation by cascades from higher levels. All these components are assumed to be exponentials, and non-linear fitting soon reaches the limits of reliable applicability, with just a few such components. Specific combinations of lifetimes and level populations can mask certain contributions or distort decay curves beyond ‘repair’ [27]. The cleanest technique to deal with cascades when trying to extract an individual level lifetime requires a measurement of the shape of all significant cascades that feed this level (besides measuring the actual decay curve), and to combine all in a single analysis. Such an ANDC (Arbitrarily Normalized Direct Cascades) analysis [28] can be automated; it involves a stepwise comparison (via differentiation and integration) of all the decay curves. In order to reduce the influence of statistical fluctuations in the raw data, the decay curves can be represented by multi-exponential fit curves (which need not have physically meaningful parameters) [29]. Nevertheless, this ANDC approach can extract reliable (accurate) level lifetimes even in specific atomic systems that suffer heavily from cascades, as do, for example, Be- and Mg-like ions. However, in a number of cases the spectroscopic equipment available does not permit the measurement of the

significant cascades (because the wavelength range needed for that measurement may be very different from the principal observations); then cascade simulation has a role (for which many examples exist), and its results - in spite of all the above caveats - may well be more accurate than those of naive multi-exponential fits.

## *2.2. Precise lifetimes*

A second general problem of atomic lifetime measurements in BFS is the time base. The actual measurement usually entails the displacement of the exciter foil relative to the line of sight of the detection system. Such mechanical displacement is easily done with high precision; however, the transformation to the time that has elapsed since excitation (more precisely: since excitation ended when the ions left the exciter foil) requires a precise knowledge of the ion velocity. Energy loss of fast ions in the foil (by collisions with the electron gas) is a statistical process; the ions of any sample may have entered the foil with basically the same velocity, but will have slightly different velocities afterwards. The actual velocity may be measured by deflecting the ion beam in a magnetic or electric field (this may be impractical at high ion energies), or approximated from the energy loss expected for the given ion species at a given incoming beam energy in a foil of given material and areal density. Evidently this estimate limits the precision of the lifetime determination at some level (and, via the Doppler effect, it also affects extremely accurate wavelength determinations - see below).

The highest lifetime precision of any classical beam-foil experiment [30] has been reached when using a time reference within the actual foil-excited ion beam, exploiting quantum beats from a well-calculated and thus precisely known fine structure interval in neutral helium. The quantum beats were observed concurrently with the light of primary interest, but through a filter for a different wavelength. Thus signal and time reference were recorded in parallel. Only very few BFS lifetime measurements have reached an uncertainty of less than 1%, and much of the limitation is caused by the problem of determining the energy loss of the fast ions in the exciter foil whose thickness is difficult to establish and which changes under irradiation.

Higher accuracy was reached when the energy loss in the foil and any geometry effect could be made small (gas excitation in beam-laser experiments [31, 32, 33, 34, 35] - although laser modulation of a foil-excited ion beam has been demonstrated, too [36]) or disregarded completely, that is, in an experiment that fed the beam of foil-excited ions into a heavy-ion storage ring [37]. There the ions were left coasting so that they passed a stationary detector on each revolution in the 55-m circumference storage ring. The actual lifetime determination consisted of straightforward electronic timing (and a small correction for the second order Doppler effect, that is, relativistic time dilation). The 10 millisecond atomic lifetime in the doubly charged (Be-like) carbon ions was thus measured with an uncertainty of 0.14%. It should be noted that the single calculation that claims an outstanding accuracy of 0.5% in this case [38] has a result that is slightly farther away from the experimental data than the combined error bars. This fraction-

of-one-percent accuracy is in the same class as reached elsewhere in fast atomic beams (produced by neutralization of a beam of singly charged ions in a gas and excitation by laser light) which can be characterized rather well. Another decade later, such high accuracy of lifetime measurements has also been reported from an electron beam ion trap [39, 40], again a measurement that avoids changes of observation geometry and that relies on electronic timing.

### *2.3. Delayed observation*

The fact that the observation zone in beam-foil spectroscopy can be positioned at the foil, at the ion beam near to the foil, or at the ion beam far downstream of the foil, offers a multitude of interesting opportunities. A short time window (high time resolution) is obviously good for the measurement of short level lifetimes, and lifetime measurements to less than 1 ps have been reported [41]. However, the decisive parameter here is not the spatial width of the detection field-of-view, but the known detail of its ‘wings’. This detail actually is what probes the true decay curve shape and determines the detail to be reconstructed from the measurement that represents a convolution of the exponential decays and the detection ‘window’. A number of such approaches have succeeded in achieving high time resolution, employing empirical window functions or synthesizing such functions on the basis of the detection geometry [42, 43, 44, 45].

Even shorter atomic lifetimes, in this case lifetimes of inner-shell vacancies in the projectile ions, have been studied by measuring the X-ray yield as a function of foil thickness [46]. Since such inner-shell vacancies are mostly produced inside the target foil, this means a measurement on the ion beam while it is traversing the solid. With spectrometers refocused for observations of the fast ion beam the Doppler broadening otherwise present has been avoided; such spectrometers have been employed to measure the line broadening that is associated with very short level lifetimes, especially of ions with autoionization decay channels [47, 48]. Observations very close to the rear surface of the foil often are swamped by a plethora of spectral lines from these short-lived core-excited states. These spectra usually are cluttered beyond analytical means. However, displacing the foil a little bit more, the time resolution offered by the observation geometry is often sufficient to suppress the light from the foil. There the observation benefits from the low-density environment and permits to record almost background-free spectra.

After excitation ends, the level populations (with the exception of levels that are temporarily replenished by very fast cascades) decrease, and so does the overall light intensity emitted by the ion beam with increasing distance from the foil. In most cases, one strives to observe the light emitted rather close to the foil, because there is most light. For certain purposes, however, it may be beneficial to observe the spectra far from the foil: exactly because the short-lived decays are diminished, the relative line intensities in spectra recorded away from the foil are shifted towards the decays of longer-lived levels. The time-integrated emissions (representing the initial level populations)



may be rather similar, but the time-differential emissions are drastically different. So in ‘delayed’ observations of overall low line intensities, the (relatively faint) spectra can be dominated by lines that are too weak to be seen in the spectra recorded near the foil where too many other spectral features mask them. In exactly this way, beam-foil EUV spectra of iron have been recorded, which close to the foil would be dominated by, for example, the resonance lines of Na- to Si-like Fe ions (typical lifetimes in the range of 10 to 200 ps). Several centimeters away from the foil (several nanoseconds after excitation) the spectra are dominated by intercombination transitions in the same ions (typical level lifetimes are in the range of 10 to 100 ns). For example, BFS observations at Bochum enabled unequivocal identification (by wavelength and lifetime measurements) of lines that had been seen in the EUV spectra of the solar corona before, but had remained unidentified. The mediocre wavelength accuracy of the accelerator-based experiment (for lack of calibration lines in the delayed spectra) was immediately improved upon by the older solar observations. This technique has been used to identify a number of further lines in the solar coronal spectra of iron group elements, and it should be able to provide many more [49]. However, because one works with delayed spectra, the light intensity is low, and a fairly intense ion beam is needed. This limits the choice of accelerators; the fully electronic Dynamitron tandem accelerator at Bochum with its high-current ion sources and wide-range accelerator capabilities was well suited for this work.

The benefit of time delay after excitation can be exploited even further, aiming at levels with lifetimes not just in the nanosecond, but in the millisecond or even second range. Traditional beam-foil spectroscopic observations then fail, because a decay length (lifetime times ion velocity) may be many kilometers, and practically no light intensity is seen in a single-pass experiment by any detection system. However, if one bends the ion beam around and lets it pass in front of the detector over and over again (the storage ring concept), then one can monitor the eventual decrease of the light intensity of electric-dipole forbidden transitions as they appear in the low-density solar corona or in even lower density planetary nebulae. The excitation process takes place before the ions (one charge state of one isotope only at a time) enter the storage ring, and for this purpose, the stripper foil or gas inside a tandem accelerator are quite appropriate. The technique may be seen as an extension of beam foil spectroscopy, doing lifetime measurements without moving a foil. Combining heavy-ion storage ring work for low-Z ions and single-pass experiments for high-Z ions, the intercombination transition rate in Be-like ions has now been studied from  $B^+$  to  $Xe^{50+}$  (see [14]), providing consistent benchmarks for theory. On electric-dipole forbidden transitions, it has been found by similar experiments that there are cases that are well described by theory, to better than 1%, and others in which calculations scatter around or deviate from experimental findings by 20 to 50%. The results (see reviews [15, 16] and ongoing research) clearly indicate where more work is needed to make reliable collisional-radiative models for plasma and astrophysics.

#### 2.4. Isoelectronic systematization

Although beam-foil spectroscopy can reach all charge states of all elements (examples are H-like U [23, 50], He-like U [51], Li-like U [52]), it would seem a waste of resources if one were trying to fill all the gaps in the data bases by experiment. Those gaps are wide, very wide, indeed, especially for elements beyond Ni ( $Z=28$ ), which are of little astrophysical interest, and with a few exceptions (Mo, Ta, W) are also of limited interest for fusion research. Experimental data, however, are needed at least in quite a few reference data cases, for example, for quality testing of atomic structure calculations and for finding out the practical ‘semiempirical’ adjustments to the results of scaled calculations. In some cases, for example in the quest for closely matching wavelengths of lines for prospective X-ray lasers based on photopumping between ion species, the needed spectral information exceeds the accuracy of any available calculations. Also, atomic lifetimes of selected levels are useful to test the predictions of radiative transition rates. It is not possible to produce a valid listing of the ‘necessary’ benchmarks that experiment has to cover. ‘Systems of interest’ are a moving target; new data needs can arise, or new measurement capabilities may move other systems into view that had been overlooked before.

The most ‘fundamental’ ionic systems (that is, those that can be calculated with the highest accuracy) are ions with only one or two electrons. Even such apparently simple ions harbour a multitude of challenges, from extremely precise energy measurements in pursuit of testing QED contributions, to higher-multipole order decays (the M1 transition in He-like ions, M2 decays, two-photon (2E1) decays in H- and He-like ions), and hyperfine-induced decays. Much of this work has been started at Berkeley in the 1970s [53, 54, 55, 56]; in the period covered here, the activities of the Argonne/Notre Dame/Toledo collaboration (Dunford, Berry, Curtis, Livingston) stand out [57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78], and GSI contributed at the high-beam energy end of the range, too (see below). (Ions with hyperfine interaction or with more electrons are discussed in the next sections). Particularly interesting results on (less heavy) He-like ions have also been reported by the Oxford/Tallahassee collaboration (Silver/Myers) [79, 80] and especially from the work on laser-induced fine structure transitions [81, 82, 83, 84, 85, 86]. Specific examples of isoelectronic sequences of ions with more than two electrons will be discussed below.

Observations of fast ion beams suffer from Doppler shift (first and second order Doppler effect) and Doppler broadening (because of the finite solid angle of observation). It is difficult to control the measurement geometry sufficiently well so that the angle between the ion beam and the optical axis of the detection system is exactly  $90^\circ$  or at least accurately known, so that a stationary lamp can be used for wavelength reference. However, if the ion beam emits light of well-known wavelengths, an in-beam calibration can achieve remarkable accuracy. Among the most precise fast-ion beam wavelength measurements are two EUV measurements. One of these measurements is of a decay in the triply excited Li ion, of the  $2p^3\ ^4S_{3/2}$  level [87], a line not very far from the

$\text{Ly}_\alpha$  transition of Li III at 13.5 nm. The other example is of the  $3s^2\ ^1S_0 - 3s3p\ ^3P_1^\circ$  intercombination transition in the Mg-like spectrum Ar VII. In second order diffraction this line practically coincides with the third order image of the well-known resonance line. When the beam-foil experimenters had (partially) resolved the line blend by varying the time delay of the observation and established the intercombination transition wavelength [88], they found out that the same had been achieved years before in a tokamak spectrum [89] - with similarly high precision. However, high-precision wavelength measurements with moving light sources like the ions of the fast ion beam are rare. Among the outstanding examples are the measurements on Li-like (and later Be-like) ions of medium-high Z, first at GANIL [90, 91] and then at GSI [92, 93, 94, 95, 96, 97]. These were later improved upon by work at the Livermore electron beam ion trap [98, 99]. The accuracy of all these data challenges atomic structure theory - not in principle, but by the accuracy of detail that the various implementations of algorithms can reproduce. The study of several ions along an isoelectronic sequence is particularly important for such high-accuracy work, because the study of various elements provides for consistency checks against technical problems such as line blends (in the lines of interest or in calibration lines) in addition to following possibly different Z-dependences of, say, QED contributions to specific level intervals. QED effects are, of course, largest in the heaviest elements. However, nuclear size effects matter, and the uncertainties of the nuclear size are smaller for some elements lighter than uranium (especially the doubly magic nucleus of Pb) - and that is why the above systematic measurements in the middle of the periodic table have a value of their own.

When thinking about where BFS in general may be heading, the bar perhaps should not be raised so high. Not all useful experiments have to be at the leading edge of what might be possible. In some cases, it would be quite sufficient to apply present capabilities to specific problems in order to be able to reap valuable insight. Other experiments might provide systematic extensions of existing data to other elements, charge states, or wavelength ranges. Let me (in the following section) consider a few topics that have found recent interest, before discussing where future work might be done in order to learn more about these topics. Lastly, I want to expand on some ideas about what to measure.

## 2.5. Relativity and hyperfine structure

Prof. Ingmar Bergström at Stockholm had taken part in the 1967 BFS conference at Tucson. He suggested to graduate student Indrek Martinson that he spend a year there, which he did, and then Martinson continued beam-foil work at Stockholm while Bergström's interests took him to CERN. On one of the visits home, the professor asked the young fil. dr. (a Swedish PhD) about what he was presently doing, and the answer was "measuring spectra and lifetimes" (of Be) by beam-foil spectroscopy. Seeing the reaction of his master that this was "nothing new", Martinson next time answered that he was "studying the effects of relativity on off-diagonal matrix elements". The clearly

delighted professor immediately prepared a letter to Prof. Stanley Bashkin in Tucson that told him the good news on interesting work that surely merited an invited talk at the next BFS conference [100]. To any practitioner in the field it should have been obvious that Martinson had changed the factual description for the advertising of the purpose, of exactly the same type of work as he had done before and as would be done by many others for several decades. After all, many measurements in physics and elsewhere consist of reading meters and indicator needles (now digitized), and the spectra and lifetimes represent the meter readout numbers and indicators on an atomic scale. Of course, accurate measuring instruments ought to be applied to interesting cases, or the effort is wasted. Detailed spectroscopy and accurate lifetime measurements are of limited value on systems that theory can describe well enough on its own (there are still occasional challenges to be met). The interesting cases are those in which theory has problems, or in which at least modern experiments can test aspects of theory that have not been accessible to testing before. Among these cases are relativistic effects that are exhibited in the drastically different decay probabilities of fine structure levels of a given term of heavy ions, and some results of hyperfine mixing that go beyond “your everyday hyperfine structure” in specific heavy ions.

Fine structure is a relativistic effect, splitting angular momentum  $L$  terms into levels of different  $J$  values and also mixing levels of similar total angular momentum  $J$  but different total spin  $S$ . Such mixing of, say, the  $J=1$  levels of  $^1P^o$  and  $^3P^o$  terms adds a decay channel, an intercombination decay. This is another of the aforementioned “off-diagonal matrix element” and will be dealt with in the next section. Besides the mixing matrix element and its variation with the nuclear charge  $Z$  (as relativistic effects increase along an isoelectronic sequence), however, there is a relativistic effect on the actual wave function, distorting the radial wave function from its non-relativistic shape. Due to this influence, there are low-charge high- $Z$  ions that feature very strong relativistic effects, and the Toledo group of L J Curtis has demonstrated this, with use of their small heavy-ion accelerator, on a wide spectrum of cases [101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120, 121, 122, 123, 124]. Recently the group has emphasized the need of obtaining not only level lifetimes (which can be converted only in unbranched decay cases to transition probabilities as are provided by theory), but also branch fractions, the ratio of an individual decay branch transition probability and the sum of all such decays of a given level [125].

Hyperfine interaction adds to the complexity of fine structure mixing for ions with a non-zero nuclear spin [126]. Both, fine structure and hyperfine structure, are considered to be well understood. In practice, however, theory still has problems to calculate fine structure intervals with satisfactory accuracy, whereas many fine structure intervals can be determined very accurately by straightforward spectroscopy or, in some cases, by laser spectroscopy. Hyperfine mixing can provide a decay channel to the otherwise extremely long-lived first excited  $J=0$  levels of two-electron ions. This effect has been demonstrated by beam-foil lifetime measurements of He-like ions some thirty years ago [55, 127, 128]. The mixing effect depends on the energy differences between the mixing levels; this

correlation can be employed to predict the ‘mixed’ lifetime from the structure. In highly charged He-like ions, however, the level lifetimes may be so short that significant level broadening spoils the spectroscopic evidence. Moreover, there are changes in the level sequence, caused by the strong ( $Z^4$ ) dependence of the fine structure splitting (in comparison to the weak (linear)  $Z$  dependence of the non-relativistic level energies in the same shell. Near to the level crossing points, the hyperfine mixing - if present - will be amplified correspondingly. One of the most precise measurements of the effect has been done on He-like Ag ( $Z=47$ ), close to such a crossing [129]. The proximity of the mixing levels combined with the lifetime broadening precluded any meaningful spectroscopic analysis; in fact, the lifetime measurement of the hyperfine mixing was then evaluated the other way round and helped to establish the level position for a systematic analysis along the isoelectronic sequence. The heaviest He-like ion for which such hyperfine measurements in the form of lifetime measurements have been made is apparently Au ( $Z=79$ ) [130].

In these He-like ions, there appears to be good agreement of calculation and measurement of the hyperfine mixing effect. Very recently, the heavy-ion storage ring has been evolving as a tool for the study of similar effects in Be- and Mg-like ions. In these ions, the resonance transition rates are lower by many orders of magnitude in comparison to the He-like ions, and thus the lifetimes that result from hyperfine mixing may be in the millisecond range instead of the nanosecond range. Otherwise there is no ‘new physics’ expected, but several measurements [131, 132] indicate significant (40%) deviations of the observed decay rates from the usual predictions. Apparently, the quality of the  $J=0$  level wave functions deserves to be improved (new, not yet published calculations differ by some 20% from the old results), and this is being revealed - somewhat unexpectedly - via lifetime measurements of hyperfine-induced decays. So lifetime measurements are still being done, more than 40 years after Prof. Bergström’s inquiry, and interesting atomic physics details are still surfacing.

## 2.6. Data on Li, Be, Ne, Na, Mg, Al, Si, Cu, Zn, Ga, Ge, Kr etc. sequences

Fast-ion beam excited foils have continued to serve as a very rich spectral light source. Spectra and atomic level lifetimes of various charge states of a given element, and spectra of various elements, but of ions of a given number of electrons, have been the staple of BFS, and have reached high accuracy in wavelength and lifetime determination, as well as high reliability in line identification, the latter process benefitting greatly from the concurrent improvements of atomic structure calculations, in particular of those that permit semi-empirical scaling referring to accepted benchmarks [133, 134, 135, 136, 137, 138, 139, 140, 141, 142].

Beam-foil spectroscopy has been thriving on the concept of isoelectronic sequences, that is, the variation of a decisive parameter, the nuclear charge  $Z$ , in an atomic system of a given number of electrons. Because the transition energies of  $\Delta n=0$  transitions scale only linearly with  $Z$  (as do their transition rates in the absence of relativistic effects),

these transitions can be studied along particularly long sections of the isoelectronic sequence. Some transitions in the one- (Li, Na, Cu, Pm) and two- (Be, Mg, Zn, Sm) electron spectra have been pursued up to U ( $Z=92$ ), though not all by beam-foil spectroscopy. With a single valence electron, it is usually an accurate transition energy of the resonance line that is wanted, whereas with two or more electrons in the valence shell, electron-electron interactions are of interest, as are intercombination transitions and their transition probabilities. The rare-gas isoelectronic sequences (He, Ne, Ar, Kr) are related to the same physics, with resonance and intercombination transitions, but here the first excitation step involves a change of principal quantum number  $n$ . Curtis has provided a large number of insightful systematizations of data along isoelectronic sequences, especially by trying to combine measured spectroscopic data (for the benefit of the multiplet mixing effects) with long range trends, and repeatedly including the calculated high- $Z$  limit in the semiempirical trend analysis. Such procedures are expected to yield most useful interpolations from very few, high-quality data points [14, 143, 144]. Unfortunately, trial and error are sometimes involved in finding out what data set should be used for the benchmarks.

Sometimes one learns more from the errors than from an eventually successful analysis, and in this spirit (having had my share in some mistakes) I am expanding a little on some of those. In the Li sequence, the lifetime measurement on  $U^{89+}$  [52] seemed to be in disagreement with the isoelectronic trend, until Curtis realized the necessity of another relativistic correction to a matrix element - and he could then show a consistent trend all across the periodic table [145]. In Be-like ions, the lifetime measurement of the  $2s2p\ ^1P_1^o$  resonance level is seriously perturbed by the cascade from the  $2p^2\ ^1S_0$  level which is only some 20% faster. There have been experimenters who failed to take this effect into account which typically results in a systematic effect of some 40%. However, quite a number of correct evaluations exist, but they scatter around the trend by much less than the individual error: apparently the estimated uncertainty of the evaluation was larger than the leftover true systematic error [14]. As mentioned before, Curtis has combined spectroscopic data and theoretical atomic structure knowledge into a mixing angle formalism that should make isoelectronic intercomparison more reliable [146]; in this approach, the spectroscopically determined level structure is combined with calculations of the multiplet mixing matrix elements. From the joint description, the analysis of isoelectronic trends and the interpolation of experimental data are made more linear and thus easier to achieve. The lifetime data on the intercombination decay ( $2s2p\ ^3P_1^o$  level) in Be-like ions can thus be presented independently of individual calculations. The high- $Z$  limit of the data points to Curtis' expectation, but the low- $Z$  data from a heavy-ion storage ring, the data with the smallest uncertainty by far, deviate from Curtis' predicted trend (see [14]). There are possibly hidden details in the wave functions that elude the otherwise intriguing mixing angle approach.

Beam-foil lifetime measurements on Na-like ions reach up to Au ( $Z=79$ ), at least for the  $3p_{1/2}$  level that is much less affected by relativistic effects than the  $3p_{3/2}$  level [147, 148, 149, 150, 151]. As in the Li sequence, the lifetime data for both fine structure

levels agree sufficiently well with expectation. A recent re-analysis of the Au spectra [152] has revealed that the 3p-3d cascade lines had been misidentified, but this had no notable effect on the interpretation of the 3p decay curves. The same re-analysis has affected the intercombination transition in the Mg-like ion of Au. In the lifetime data of this sequence, there is a notable oddity. Many lifetime data, from ion trap and storage ring measurements at low  $Z$  to beam-foil experiments at medium and high  $Z$ , agree with the expected trend of some veteran relativistic calculations [153]. One series of experiments, performed by a Swedish-Japanese collaboration at a RIKEN accelerator, however, branches off towards higher transition rates. Their data carry small error bars and are mutually consistent [154], but they follow a clearly different trend with a slope that is incompatible with that of either established theory or the other published data on this sequence. The experimental set-up at RIKEN has been taken apart and therefore the measurement can't be repeated there.

The same spectra that contain the intercombination lines of the Mg-like ions also contain the nearby intercombination transitions in Al- and Si-like ions, which represent the same physics, but may be even more challenging to theory, because of the interaction of several electrons in the valence shell (and not to forget other E1 transitions in these ions, see, for example, [155, 156, 157, 158, 159]). Especially the decay of the  $3s3p^3\ ^5S_2^o$  level in the Si-like ions is strongly affected by interference effects of triplet levels; observations and calculations had problems in the middle of the elemental sequence. Recent calculations of a higher accuracy than available before have enabled a re-analysis of earlier beam-foil data for Xe and Au [152, 160], and in combination with spectra from the electron beam ion trap at Livermore, the line identification problems appear to have been solved.

Rare gases and rare-gas sequence ions have shown various surprises in atomic structure and particularly in atomic lifetime studies. When Curtis tried to systematize the Ne sequence, he found in the literature, for example, three mutually exclusive lifetime data on neutral Ne, Bochum beam-foil data for  $\text{Na}^+$  [161], a few beam-foil data on Cl and Ar, and data on the resonance and intercombination decay rates in Ne-like S and Cl [162] from Lund that carried exceptionally small error bars. Unfortunately, the combination of the few high-quality data and the expected high- $Z$  limit did not make much sense, and the less precise data just seemed to scatter wildly. I was asked to remeasure the Na data of old and to measure the same levels for as many elements as possible. Establishing the role of systematic errors by cascades for some elements, lifetimes up to S ( $Z=16$ ) were obtained [163], not as precise as in the two-element work reported from Lund, but with a different isoelectronic trend that disagreed with the Lund data by some 6 to 7 of their standard deviations. Eventually the Lund group revised their data for S [164], and their new values come closer to the isoelectronic trend of the other data and the high- $Z$  limit predicted by Curtis, but there is certainly room for improvement. In Ne-like ions, the resonance lines (opening the  $n=2$  shell) have much shorter wavelengths than the transitions in the  $n=3$  shell, and usually require a different spectrometer for the observations. Nevertheless, the Lund group has managed to determine precise absolute

excitation energies of Ne-like ions without observing the resonance lines [165].

Cl-like (as well as Al-like) ions of iron group elements feature prominent forbidden lines in the solar corona and comparable astrophysical light sources. Their rates are too low for ordinary BFS work (but they are being measured in heavy-ion storage rings and electron beam ion traps [166]). However, Cl-like ions also have a displaced term in the  $n=3$  shell,  $3s3p^6\ ^2S_{1/2}$ , with a resonance line doublet decay to the fine structure levels  $3s^2\ 3p^5\ ^2P_{3/2,1/2}^o$  of the ground term. The two lines are readily observed in many plasmas, including the sun. One might naively assume that this lowest excitation, far from the  $n=4$  shell and any level crossings, has been treated by theory long since and sufficiently well. Indeed, it has been treated often, but a beam-foil study at Bochum found a considerable scatter of the predictions for the level lifetime as well as for the branch fractions, and not many of the calculations were compatible with the results of measurements on Fe, Ni and Cu [167, 168]. At the low-Z end of the sequence, the lifetime predictions differed from the few (not very precise) measurements and from each other by factors of five and more. New calculations [169, 170] came much closer to the BFS data, but a new measurement (exploiting synchrotron light to photoionize Ar) [171] set a new, accurate benchmark that eluded those more recent calculations. A dedicated calculation for Ar II [172] then came close to the good data point, but, alas, it does not cover any ions further along the isoelectronic sequence.

In many aspects, multiply charged Cu-like ions can be considered ions with a single  $4s$  electron outside a closed  $3d^{10}$  shell, a relative to the alkalis. However, if the  $3d$  shell is opened, as it is easy to do in the intense beam-foil interaction, there appear  $3d^9\ 4s^2$  and  $3d^9\ 4s4p$  levels, which resemble the alkaline earths with some added complexity due to the open  $3d$  shell. There are long-lived levels which play a role in some x-ray laser photopumping schemes, and BFS has been put to good use in providing data on the decays of long-lived levels [173, 174, 175].

The Zn isoelectronic sequence should be in many ways similar to the Mg sequence, with resonance and intercombination transitions and rather the same problems of specific cascades feeding the resonance line. Even with the presence of such tricky problems thus known in advance, it has taken several iterations before experiment and some of the calculations found common ground [176, 177, 178, 179, 180, 181]. A very recent development is the exploitation of precisely measured or calculated polarizabilities to derive certain resonance transition line strengths (and thus transition rates); this new approach makes it possible to revisit a number of older beam-foil lifetime results that suffered from spectral blends or had unresolved discrepancies among the results obtained by various groups [182, 183]. Just like the Al- and Si-like ions have lines that fall close to the strong lines in Mg-like ions, the significant lines in Ga- and Ge-like ions appear in a similar pattern near those of the Zn-like ions, and the same pattern is expected to repeat for Pm-, Sm- and Eu-like ions. For this reason, many of the beam-foil studies of such ions cover three charge states of a given element at a time [184, 185, 186, 187, 188].

In the Kr isoelectronic sequence, beam-foil spectroscopy has traced how the lifetime of the  $4d\ ^3P_1^o$  level has a counter-intuitive trend of lifetimes increasing with the nuclear



charge [189]. The trend was confirmed by calculations, but no simple argument was available to explain this behaviour.

### *2.7. Iron group element EUV spectra for astrophysics*

In astrophysical observations of remote objects, low-charge state spectra dominate. Such low-charge state ions can be produced in a great many light sources, but the astrophysical environment is one of very low particle density, and the spectral patterns of emitters under low and high density conditions differ considerably. The emission of long-lived levels - often by electric-dipole forbidden radiation - is easily quenched by collisions under any density that is not what we call ultrahigh vacuum (UHV). Most spectroscopic work in the laboratory has been done at much higher densities (atmospheric pressure and down to one thousandth or a tenthousandth of that, the typical conditions of gas discharges). It should be worthwhile to actually employ a low density light source like an electron beam ion trap (with a low energy electron beam) in a systematic spectroscopic survey of at least the optical and near-IR ranges in order to obtain spectra with relative line intensities as encountered in astrophysical observations. Beam-foil spectroscopy cannot do the same, because the excitation takes place under extremely high-density (solid state) conditions. The aforementioned work at Toledo that concentrates on low charge ions therefore is not aimed at spectroscopic surveys, but at detail studies of the atomic structure and dynamics. Similarly, that extension of BFS to heavy-ion storage rings is concerned with measuring (intercombination and E1-forbidden) decay rates also of low- to moderate-charge ions under UHV conditions (see [15, 16] and references therein), but the high velocities of the stored ions cause considerable Doppler shifts which are difficult to take care of in precision spectroscopy, and the storage of individual ion species makes for clean observation conditions, but would be extremely tedious for survey work. Mannervik has reviewed the early years of low-charge state ion studies using heavy-ion storage rings [190].

However, BFS can and does play a role in the understanding of astrophysical spectra beyond the demonstration experiments that marked the beginning of the field [2]: From our celestial light source, the sun, we can detect and disperse light also in the EUV and x-ray ranges. This light is largely emitted by highly charged ions in the solar corona, and the decay rates are commensurate with BFS and its derivative techniques. In fact, owing to the fact that the solar light source has the usual elemental abundances, plus ions of each element in various charge states, there are very many emission lines. A recurring theme in many reports on solar spectroscopic work based on sounding rockets or satellite observations is that about half of the observed lines remain unidentified. Of the identified lines, of course, the shortage of laboratory data at the time and the ubiquity of line blends work against a complete and fully unambiguous analysis, and errors of assignment must have occurred. It is time for a systematic re-analysis of the solar EUV spectra, starting with the spectra of Fe (because of its high abundance) in the middle charge states (most prominent under coronal temperatures (a

few hundred eV)). Such analysis combines terrestrial, solar and extrasolar observations (by the *Chandra* and *XMM – Newton* spacecrafts) with extensive atomic structure and electronic excitation calculations in order to obtain synthetic spectra [191] that then can be compared to observations [192, 193].

It is a major effort to combine the data from such very disparate sources, and calculations of spectra excited under different conditions are required to judge the observations at a given light source and their relation to solar observations. BFS observations usually do not reach the spectral resolution of solar observations or of measurements at tokamaks, and they result from high-density excitation, but they have the great advantage of relating to a single isotope, of a narrow range of charge states, and of time resolution that can distinguish between fast and (moderately) slow emitters. In this spirit some BFS observations at Bochum have helped identify various intercombination transitions in Mg- to Si-like ions of Fe and neighbouring elements [49, 194, 195, 196, 197, 198]. Delayed spectra (the observation window being displaced from the exciter foil along the ion beam trajectory) helped to suppress emission from the plenitude of core-excited states that are reached only under high-density conditions, making the decays of long-lived levels better recognizable. Lifetime measurements on the more prominent lines served to corroborate line identifications (and to check on calculations); on weaker or partially blended lines, the decay curves helped at least with the identification process. However, some of the Fe lines seen in the solar corona have not been seen in any laboratory light source yet. The maximum foil displacement in the Bochum measurements corresponded to less than 20 ns after excitation, but some of the levels of interest have predicted lifetimes of hundreds of nanoseconds (some intercombination transitions) or even milliseconds. This renders the signal per path length interval observed too low to distinguish from background.

Some of the extremely long level lifetimes were then measured at the Heidelberg heavy-ion storage ring, of 3p and 3d levels in FeX through FeXIII (and in some neighbouring elements) [199, 200, 201, 202, 203], and in FeXIV at the Livermore and Heidelberg electron beam ion traps [204, 40]. The measurement accuracies of one percent and better clearly elucidate which of the available calculations yield reliable values for the E1-forbidden transition rates between ground configuration levels. These transitions modify the relative populations of those levels from which electronic excitation starts, and thus the lower level populations and their dynamic balance of excitation and radiative decay renders specific lines to reflect the particle density in the plasma. Benchmark data of the ground configuration levels can then be used to tie down key parameters in the spectral modeling effort. The most prominent of the E1-forbidden transitions in, for example, Al- and Cl-like ions of Fe, have wavelengths not in the EUV, but in the UV and visible spectral ranges, and they are not only seen in the solar corona, but in a wide variety of astrophysical objects. It is amazing to see how information from across the universe can be gathered and understood by some atomic physics measurements done under ultrahigh vacuum conditions - by techniques that are offsprings of or closely related to BFS!

### 3. Migration

Formerly, most BFS work was done in North America and Europe. However, of the American and Canadian accelerator-based atomic physics laboratories in Tucson (AZ), Berkeley (CA), Argonne (IL), Notre Dame (IN), Toledo (OH), Oak Ridge (TN), Knoxville (TN), College Station (TX), Tallahassee (FL), Brookhaven (NY), Edmonton (AB), London (ON), and Laval (Québec), and their European counterparts in Sweden (Uppsala, Stockholm, Lund), Denmark (Aarhus), Great Britain (Manchester, Oxford, Daresbury), France (Lyon, Grenoble, Strasbourg, GANIL/Caen), Belgium (Liège, Louvain-la-Neuve, Bruxelles), The Netherlands (Groningen) and Germany (Heidelberg, Kaiserslautern, GSI/Darmstadt, Giessen, Munich, Berlin, Cologne, Bochum), not much is left with any notable BFS activity (the above listing is bound to be incomplete, both by my faulty memory and by other people's possibly different view of what would qualify to be included). The Japanese effort at Tsukuba, Kyoto, RIKEN and at some smaller laboratories should also not go unnoticed, but the latest BFS effort, at RIKEN, has ended recently. Heavy-ion accelerators have been built in other countries, too, but nothing like BFS apparently has been pursued in the Soviet Union / Russia, Argentina, or Brazil. India has several high-tech laboratories in the Mumbai (Bombay) area, and under S K Mitra the accelerator laboratory at Tata Institute (TIFR) was evolving nicely (for example with forefront beam-foil work [205]) - until the head died, and atomic physics activities apparently closed down. There is also an accelerator center (Nuclear Science Centre NSC) at New Delhi (with a 15 MV tandem accelerator) which would seem capable of following up on many of the earlier American and European investigations. Indrek Martinson at Lund has run the beam-foil group at Lund as a scientific hospitality center for several decades. There has been spectroscopic beam-foil work from Beijing (P R China) in which Martinson and Christer Jupén have played the role of conveying expertise [139, 140]. At Beijing and at Lanzhou (heavy-ion storage ring laboratory) accelerator-based atomic physics is now established, and some of the experience gained in the USA and Europe will continue there.

Unfortunately, the migration of knowledge is more complex and difficult than setting up similar equipment in another place. Large scale equipment is not essential to do good work in science. In some laboratories, rather small accelerators are available as leftovers from early days, and even groups in some developing countries might be able to secure affordable second-hand machinery (for example, implanters from semiconductor industry) and run the equipment. Because of all the the earlier work in the field, the remaining BFS research opportunities with such machines are limited nowadays, but some productive research work, especially when training young researchers, is still possible in niches. For example, in the Toledo accelerator laboratory Larry Curtis has trained a string of graduate students on mostly low-charge heavy ion projects; in some of these ions there are massive relativistic effects, for example, large lifetime differences between fine structure levels. Such valuable atomic physics work could well continue elsewhere. For example, there are results from the small machine at TIFR

(Mumbai) [206, 207] that make contributions to low-energy accelerator-based atomic physics. However, it is essential that researchers (and journal referees) be aware of past work (and past mistakes). There is no merit in repeating past experiments with poorer means.

#### 4. Opportunities

In this section I discuss some personal ideas of what BFS can do best and should be used for. These problems are stretching the boundaries of what can be done by BFS in various ways that I shall explain. They address specific problems of atomic structure, of peculiar decay modes, or relate to astrophysics. Maybe I should emphasize that this is a selection based on my personal preferences; there are, for example, many unsolved problems in ion-matter interaction, but the following examples only deal with atomic structure and dynamics. Even here, what I presently see as deserving to be tried, has changed over the last few years. For example, more than a quarter of a century ago, I have recorded high-resolution EUV beam-foil spectra of Si in the wavelength range from 10 to 20 nm. These spectra contained mostly 3 - 4 transitions of Li-like, Be-like, and B-like ions of Si. The 3d-4f lines in the Li-like ion were known at the time from laser-produced plasmas, and educated guesses could be made on some of the 3d-4f transitions in the other ions. In fact, such yrast transition multiplets are most prominently excited in BFS and yield access to some interesting structure detail. However, it took several attempts until more than a decade later that the bulk of the lower- $\ell$  transitions in the Be-like part of the spectrum could be reliably disentangled [208], and more than another decade to reach the same for B-like Si [209]. Now, however, calculations, such as demonstrated by Vilkas and Ishikawa on Si X, can be used for many other cases, reducing the need to measure most of the spectra - and enabling unprecedented checks on line identifications made before such consistency checks were available; some inconsistencies may even appear in the critical work listed in the NIST spectral tables! In fact, such calculations have recently been applied to a re-investigation of some of our Xe and Au spectra obtained at the UNILAC accelerator of GSI Darmstadt more than ten years ago. The new calculations represent such a massive improvement in accuracy that several line assignments could be corrected [152, 160]. An estimate of the overall wavelength accuracy of these calculations in the EUV range, of about 0.005 nm, is borne out by the comparison with better resolved spectra from a stationary light source, the (Livermore) electron beam ion trap. That low-density light source and the beam-foil light source complement each other [210], one providing benchmark wavelengths on a few ground state transitions, the other adding a plethora of lines that are excited only under high density conditions. The latter aspect is what is of interest here. Moreover, BFS offers unique observation with time resolution on the picosecond and nanosecond scale.

#### 4.1. Core-excited levels

The ion-foil interaction is very effective for reaching high-spin term systems and multiple excitation. One example is the optical observation of the decay of core-excited  $\text{He}^-$  ions and their isoelectronic counterparts up to  $\text{C}^{3+}$  [21, 22]. Another example is the production of  $\text{Li}^-$ , an ion that is reasonably stable only in a core-excited state [18], and that is the first negative ion for which transitions between excited (quintet) states have been recognized and the level lifetime measured [19]. In the foil-excited Li spectrum, the  $\text{Li}^-$  quintet line is remarkably strong (and the line had been seen - just not recognized - much earlier). The isoelectronic sequence has been followed towards higher Z elements (see [20, 211]), although the UV line intensities decrease due to competition by non-radiative decays (autoionization). Weak indications of the lines have been seen for  $\text{Mg}^{8+}$ , from an experiment with the Bochum tandem accelerator and a less than optimum grazing incidence spectrometer. A fast spectrometer for the wavelength range near 50 nm, that is, near the short-wavelength cut-off of normal incidence optics, would lead to improvement.

In the same vein, studying the competition of UV and soft-X-ray radiative and non-radiative decays, are lifetime measurements of the  $1s2p^2\ ^4P$  levels of Li-like ions. At low Z, the level lifetime is dominated by the in-shell UV branch; with increasing nuclear charge the X-ray and autoionization decay branches gradually predominate. By level lifetime measurements that individually only reflect the sum of all three decay rates, it is nevertheless possible to disentangle the different contributions [211]. Apparently the atomic structure codes employed at the time underestimated the in-shell transition rates by some 20%, and similarly for the  $\text{Li}^-$  sequence. In the  $\text{Li}^*$  sequence, lifetime data have been published on  $\text{Mg}^{9+}$  and  $\text{Al}^{10+}$  [212], but data on  $\text{Si}^{11+}$  would also be valuable. Preliminary data obtained at Notre Dame have been presented in a conference abstract long time ago [213], but they have not been published formally. More complex core-excited few-electron ions have been studied by experiment in combination with theory, leading to quintet and sextet levels [214, 215, 216, 217, 218, 219].

Most core-excited ions autoionize rapidly. However, there are interesting exceptions, which usually owe their longevity to a high total angular momentum quantum number. An example in Li-like ions is the  $1s2p3d\ ^4F_{9/2}^o$  level which at moderate and high values of Z has  $2s\text{-}3d$  E2 transition (VUV) and  $1s\text{-}2p$  M2 transition (XUV) decay branches that are stronger than the E1 decay branches to  $1s2p3p\ ^4D_{7/2}$  or  $1s2s3d\ ^4D_{7/2}$  [220]. For low-Z elements, the existence of the long lived level has been demonstrated in interesting multi-step electron capture experiments [221, 222]. The experimental challenge when searching for a photon signal is the fact that there is a single long-lived level with a single emission line in each of those spectral ranges. It is difficult to identify a single line; in this case, the signature is the level lifetime of several nanoseconds for Li-like ions of, say, Si, P, or S. The soft-X-ray branch near 3 nm falls into a range with many lines of ions of neighbouring charge states. Considering typical spectra and detectors, the best chance might be in the VUV, employing a position-sensitive detector that covers a

certain section around the predicted wavelength [220], and then recording sets of delayed spectra.

Experiments using beams of relatively slow highly-charged ions from an electron cyclotron resonance ions source (ECRIS) and using electron capture from a gas have also been reported from Uppsala [223, 224, 225] and from Caen [226, 227]; such work has opened many interesting new directions, e. g. in the context of understanding the soft-x-rays seen from comets. (A type of collisional spectroscopy that is related to BFS, using a target gas instead of a foil, has been extended to very high ion beam energies at the fragment separator of GSI Darmstadt's system of accelerators [228], and the internal gas target in their heavy-ion storage ring ESR serves the same purpose.)

Another set of doubly-excited high-J levels is found in Na-like ions, with one of the 3s electrons removed to a 3d position. The high total angular momentum prohibits rapid autoionization, so that 3s-3p radiative transitions can be observed. The strongest such line in Na-like ions of elements K through Cu has been readily identified [229]. However, there are more such lines in the spectra of Na-like ions [230] and probably also of Mg- and Al-like ions. In core-excited Cu-like ions, sextet levels have been predicted and searched for by BFS [231]. Measurements would test the capability of atomic structure codes to predict not just the vast multitude of transitions in open shell systems, but to filter out and identify those few that have a high VUV fluorescence yield.

#### *4.2. Intercombination (spin-forbidden) and electric-dipole forbidden transitions*

Straightforward spectroscopy of foil-excited ion beams suffers from the excitation of too many lines that can make identification difficult in the absence of high quality calculations. However, this drawback and the often considerable line width (unless refocusing techniques are employed [232, 233, 234, 235, 236]) are offset by the fact that the BFS light source is monoisotopic: with the exception of some few carbon UV lines (from the foil, possibly after sputtering), all the light is that of a defined element, and even the charge state can be determined within a few units by suitable beam energy variation. The accumulation of a 'complete' spectral data base of all elements and from the X-ray range to the visible or even to the near infrared would be of great scientific and applied science value, but, alas, it would also be prohibitively expensive in terms of accelerator time and manpower. Any section of such a large scale project that would be tackled experimentally should be preceded and accompanied by calculations of the standard achieved by recent work in several places, by techniques such as Relativistic Configuration Interaction (RCI), Many-Body Perturbation Theory (MBPT) - especially of the Multi-Reference MBPT (MR-MBPT) Møller-Plesset type pursued by Ishikawa and his group at the University of Puerto Rico (see [209] and references therein).

Having found intercombination transitions in 12- to 14-electron ions in the delayed beam-foil spectra of iron group elements, it is suggested to follow these and similar isoelectronic sequences towards higher nuclear charges in order to determine reliably the isoelectronic trends and to see the growing influence of relativity. Also, at high

Z, a reordering of the level sequence takes place because of the transition towards jj coupling. This has the effect of certain groups of levels of low-Z displaced terms or low-Z ground configurations becoming unrecognizable at high Z [152]. Such reordering is to be recognized with the help of calculations, but detailed tests of the quality of the predictions require good measurements. Also, with increasing nuclear charge, the transition rates of intercombination (spin-changing) transitions become high enough for convenient (delayed) spectroscopy and lifetime measurements. At the highest nuclear and ionic charges, electric-dipole forbidden transitions, as are seen with moderately charged ions in many low-density plasmas (terrestrial and astrophysical), should be visible also with foil-excited ion beams.

Besides production work on spectral surveys and key level lifetimes, there are detail problems that can involve intricate atomic structure theory. One example is the  $3s^23p3d\ ^3F_4^o$  level in Si-like ions. This level cannot decay by electric dipole transition (there is no lower-lying  $J=3$  level of opposite parity), but by a multitude of M1 and M2 decay branches, all of which have rather low transition probabilities at low Z. No individual decay of this level has ever been identified. Although no individual decay channel of the  $^3F_4^o$  level is known experimentally, a storage ring experiment in which - for lack of spectral discrimination - all EUV decay channels of this level (in  $Fe^{12+}$  ions) were seen at the same time (when ions of only a single isotope and charge state were stored in a storage ring) has measured the total decay rate and has found reasonable agreement with theory [203, 237].

The neighbouring fine structure level of the same term,  $3s^23p3d\ ^3F_3^o$ , is also long-lived, but less so. A major decay branch of that level has been observed in delayed beam-foil spectra of several iron group elements [49], correcting semi-empirical estimates about the term position. The newly established excitation energies were later found to agree with the results of independently performed MR-MBPT calculations [237]. However, this single observation would benefit from work on a few other ions to tie down the isoelectronic trends of the competing decay channels.

#### 4.3. Line ratios

In the same Si-like ions (and, correspondingly, in C-like ions), the lowest quintet level,  $3s3p^3\ ^5S_2^o$  ( $2s2p^3\ ^5S_2^o$  in C-like ions), is of special interest (as has been mentioned above). The multiplet mixing that permits this level to decay towards two fine structure levels of the  $3s^23p^2\ ^3P$  ground term is not just with  $3s3p^3\ ^3P_2^o$ , but also with  $3s3p^3\ ^1D_2^o$ , and there are some strong cancellation effects with  $3s^23p^2\ ^3P - 3s3p^3\ ^3P_2^o$  transitions [9, 238]. The lifetime of the quintet level in C-like ions has by now been measured in  $N^+$  and  $O^{2+}$  [239, 240]. However, interesting as these lifetime measurements are in comparison to theory, there is another probe of the detail of the wave functions involved, and that is the branching ratio of the two intercombination lines. For the C sequence  $2s2p^3\ ^5S_2^o$  level, theory [9, 238] predicts the  $2s^22p^2\ ^3P_{1,2} - 2s2p^3\ ^5S_2^o$  branching ratio to vary from about 3 at low Z to about 1 at high Z. Low-Z experimental data see (see [241]) are

contradictory; a possible high-Z data point for Fe ( $Z=26$ ) in the solar EUV spectrum suffers from a blend of the FeXXI lines with FeXIV. This blend could be avoided in BFS by going to an ion beam energy of 150 MeV or more, as would be available (among other places) at New Delhi or at Beijing. In FeXXI, the quintet level lifetime is of order 16 ns (in Ni, 6 ns). There is no need for an accurate absolute wavelength measurement of the two lines, since it is the intensity ratio that matters most, but the wavelength difference needs to be ascertained for identification purposes. A single (out of two needed) possible candidate line has been seen in Ag spectra recorded at GSI Darmstadt in 1997/98 (unpublished work in the context of [94]), but without the other line, this demands further identification. Thus some systematic work would be required to first confirm the isoelectronic trend of the wavelengths. Unfortunately, the spectrometer and beam-foil set-up used at GSI have since been decommissioned.

Fine structure splittings increase with  $Z^4$ , whereas the in-shell energy intervals increase only linearly with  $Z$ . This is one of the reasons of the reordering of the level sequence in high-Z ions. For example, in B-like low-Z ions, there is a  $2s^22p\ ^2P^o$  ground term with a small doublet fine structure splitting. Among the  $2s2p^2$  displaced terms of the same ion, there is also a  $^4P$  term with three fine structure levels. Hence at low  $Z$  there are  $2s^22p\ ^2P^o - 2s2p^2\ ^4P$  intercombination transitions that are difficult to resolve. At high  $Z$ , in contrast, the fine structure splittings are so large that the lowest quartet level in B-like Xe lies at a position about halfway between the  $2s^22p\ ^2P^o_{1/2,3/2}$  levels, and the upper level ( $J=3/2$ ) of what used to be the low-Z ground state has two decay branches, the M1 (with E2 contributions) decay to the true ground level ( $J=1/2$ ) as well as an E1 spin-changing (intercombination) decay to  $2s2p^2\ ^4P_{1/2}$ . That level in turn can decay by a similar transition to the ground level. So in addition to the M1 transition with a wavelength near 3 nm there are two lines with wavelengths near 6 nm. Near 3 nm, the spectrum has many lines from the electric-dipole forbidden fine structure transitions in the ground configurations of various charge state ions, and it is difficult to disentangle the spectrum. Near 6 nm, the spectrum is much cleaner, and a measurement with an electron beam ion trap has, indeed, shown the lines of interest [98]. Lifetime measurements on the two lines (in the nanosecond range) in comparison to calculations would test the latter and would yield desired information on the branch fractions.

Line ratios in some cases depend on details of the wave functions that may change drastically with the nuclear charge along an isoelectronic sequence, because of level crossings or level compositions. Particularly notable examples occur in the displaced levels of B- and Al-like ions. BFS has been used to study this effect [242, 243]; the predicted trends were clearly seen, but the limited agreement of theory and experiment in detail leaves much room for more detailed work.

#### 4.4. Very-long lived levels

Beam-foil spectroscopy has been fruitful in providing lifetimes of excited atomic levels in the range from tens of femtoseconds (inner-shell vacancies, measured by foil-thickness



variation) to many nanoseconds. In the range up to a few dozen nanoseconds, BFS can help with the elucidation of solar coronal spectra by both delayed spectra and lifetime measurements on iron group elements, following the examples shown in [49]. Beyond a few hundred nanoseconds, however, the decay lengths (level lifetime times ion velocity) are in the unwieldy range of several meters and more. Ways out of this dilemma are possibly provided by slow beams of highly charged ions, as are available from recoil ions (extracted from a gas under high-energy, high-charge ion bombardment) [244] or from a suitably biased electron cyclotron resonance ion source. This technique should be workable with lifetimes in the microsecond range, preferably employing photo-ion coincidence measurements for best signal-to-noise. A good example of atomic systems to be studied this way are the intercombination transitions in Be- through C-like ions of Ne, and the corresponding transitions in Mg- through Si-like ions of Ar. All of these charge states are easily produced in the above devices. Lifetimes in the millisecond and second range are already investigated at heavy-ion storage rings such as CRYRING Stockholm or TSR Heidelberg, or employing electron beam ion traps. However, the storage ring experiments are presently not equipped to provide spectral analysis in the EUV range, and for the electron beam ion traps (which offer excellent spectroscopic coverage) time-resolving spectroscopic equipment has to be developed.

#### 4.5. Electric dipole allowed transitions

Electric dipole allowed transitions are the most ordinary of all transitions - surely all there is to know about those must be known after more than four decades of beam-foil lifetime measurements? Well, even in seemingly simple atomic systems there may be hidden complexities. Few-electron ions such as of B have been studied for decades (also by BFS), and still there are new insights from improved measurements [246, 247], and the work on B continues. One of the key achievements of BFS has been differential lifetime measurements, that is, the recording of decay curves of individual fine structure levels of certain terms. The slight differences of the slopes of (largely) straight line logarithmic plots of such decay-curve data show the presence of additional decay channels. Good examples are the observations of  $1s2s\ ^3S_1 - 1s2p\ ^3P_{0,1,2}^o$  transitions of He-like ions that revealed the growing importance of the intercombination decay of the  $J=1$  level [245], and similarly the  $2s3s\ ^3S_1 - 2s3p\ ^3P_{0,1,2}^o$  transitions of Be-like ions. The results for the intercombination transition rate in Be-like ions up to Ne show the expected trend - why continue along the isoelectronic sequence? There are  $2p3s$  levels (same symmetry!) close to the  $2s3p$  levels, and this proximity affects the wave functions significantly. Various calculations have produced very different results. Although more recent calculations appear to be more reliable than some of the old ones, experimental validation would be welcome. Such validation, not under optimum experimental conditions, has been tried for Mg at Lund and for elements Na through Si at Bochum (seeking out the  $2s3s - 2s3p$  transitions). It was tried also to measure the decay behaviour of the  $2s^2\ ^1S_0 - 2s3p\ ^3P_1^o$  intercombination directly, a measurement that yields only the sum of intercombination

and in-shell transition rates (whereas the differential measurement permits to extract both rates individually). The results were not quite satisfactory [143, 248]. A new experiment ought to combine a good ion source and accelerator with a spectrometer that has high detection efficiency in the wavelength range 100 to 160 nm. An appropriate experiment is envisaged at Bochum.

Such a spectrometer would be very interesting also to provide laboratory comparison data for the SOHO spacecraft SUMER instrument and its working range from about 50 to 160 nm. Almost half of the lines seen there are still unidentified [249, 250], and beam-foil spectroscopy can very likely supply elemental and charge state identifications for many of them.

## 5. Conclusion and outlook

Beam-foil spectroscopy and its sibling techniques have been outstandingly productive tools of atomic physics, providing both important data and insight. For some forty years, the developments have led to improvements in working range and reliability, and catalogues of desirable further measurements can be formulated. However, most of the key persons who have carried out and directed much of the development effort are nearing retirement, and with them the leading facilities. It is thus not likely that many of the desirable BFS projects discussed will presently be pursued. High-Z element, high-charge state spectroscopy and some specific long-lived level lifetime measurements will, however, be taken over by electron beam ion traps, and heavy-ion storage rings will contribute some important benchmark measurements on electric-dipole forbidden or hyperfine-induced transitions. Beam-foil spectroscopy can still be expected to solve a number of interesting atomic physics questions, but as a technique, at present, it has dropped from fashion and support.

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